Vortex lattice structures of Sr_2RuO_4

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The vortex lattice structures of Sr_2RuO_4 for the odd parity representations of the superconducting state are examined for the magnetic field along the crystallographic directions. Particular emphasis is placed upon the two dimensional representation which is believed to be relevant to this material. It is shown that when the zero-field state breaks time reversal symmetry, there must exist *two* superconducting transitions when there is a finite field along a high symmetry direction in the basal plane. Also it is shown that a square vortex lattice is expected when the field is along the c-axis. The orientation of the square lattice with respect to the underlying ionic lattice yields information as to which Ru $4d$ orbitals are relevant to the superconducting state.

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The oxide superconductor Sr_2RuO_4 is structurally similar to the high T_c materials but differs markedly from the latter in its electronic structure [1]. In particular, the normal state near the superconducting transition of Sr_2RuO_4 is well described by a quasi-2D Landau Fermi liquid [2]. There now exists considerable evidence that the superconducting state of Sr_2RuO_4 [1] is not a conventional s-wave state. NQR measurements show no indication of a Hebel-Slichter peak in $1/T_1T$ [3], T_c is strongly suppressed by non-magnetic impurities [4], and tunneling experiments are inconsistent with s-wave pairing [5]. While these measurements demonstrate that the superconducting state is non s-wave, they do not determine what pairing symmetry actually occurs in this material. The determination of the pairing symmetry in unconventional superconductors is a notoriously difficult problem and theoretical insight provides a useful guide. The observations that the Fermi liquid corrections due to electron correlations are similar in magnitude to those found in superfluid ^{3}He and that closely related ruthenates are itinerant ferromagnets have led to the proposal that the superconducting state in Sr_2RuO_4 is of odd-parity [6]. Even with this insight there still remain five odd-parity states that have different symmetry - all of which have a nodeless gap and therefore similar thermodynamic properties [6]. Recently, μSR measurements indicate that a spontaneous internal magnetization begins to develop at T_c [7]. The most natural interpretation of this magnetization is that the superconducting state *breaks* time reversal symmetry ($\mathcal{T}$). This places a strong constraint on the pairing symmetry in Sr_2RuO_4 since it implies that the superconducting order parameter must have more than one component [8]. Of the possible representations (REPS) of the D_{4h} point group only the two dimensional (2D) Γ_{5u} and Γ_{5g} REPS exhibit this property. Of these two the Γ_{5u} REP is the most likely to occur in Sr_2RuO_4 due to arguments of Ref. [6] and the quasi-2D nature of the electronic properties. The order parameter in this case has two components (η_1, η_2) that share the same rotation-inversion symmetry properties as (k_x, k_y) [8]. The broken $\mathcal{T}$ state would then correspond to $(\eta_1, \eta_2) \propto (1, i)$.

I investigate within Ginzburg Landau (GL) theory the vortex lattice structures expected for the odd-parity REPS of the superconducting state; focusing mainly on the Γ_{5u} REP. It is initialy shown that a general consequence of the broken $\mathcal{T}$ state described above is that in a finite magnetic field oriented along a high symmetry direction in the basal plane there will exist a *second* superconducting transition in the mixed state as temperature is reduced. The observation of such a transition would provide very strong evidence that the order parameter belongs to the Γ_{5u} REP. The form of the vortex lattice for a field along the c-axis is then investigated for both the one dimensional (1D) and the 2D Γ_{5u} REPS of the superconducting state. It is shown that a square vortex lattice is expected to appear for all the REPS, however observable differences exist between the 1D and the 2D REPS. Finally, within the recently proposed model of orbital dependent superconductivity of Sr_2RuO_4 [9] it is also shown that the orientation of square vortex lattice with respect to the underlying crystal lattice dictates which of the Ru $4d$ orbitals give rise to the superconducting state.

To demonstrate the presence of the two superconducting transitions described above consider the magnetic field along the $\hat{x}$ direction (x is chosen to be along the basal plane main crystal axis) and a homogeneous zero-field state $(\eta_1, \eta_2) \propto (1, i)$. In general the presence of a magnetic field along the $\hat{x}$ direction breaks the degeneracy of the (η_1, η_2) components, so that only one component will order at the upper critical field [*e.g.* $(\eta_1, \eta_2) \propto (0, 1)$]. As has been shown for type II superconductors with a single component the vortex lattice is hexagonal at T_c and the order parameter solution is independent of x so that σ_x (a reflection about the $\hat{x}$ direction) is a symmetry operation of the $(\eta_1, \eta_2) \propto (0, 1)$ vortex phase [10]. Now consider the zero-field phase $(\eta_1, \eta_2) \propto (1, i)$, σ_x transforms $(1, i)$ to $(-1, i) \neq e^{i\phi}(1, i)$ where ϕ is phase factor. This implies that σ_x is *not* a symmetry operator of the zero-field phase. It

follows that there must exist a transition in the finite field phase at which η_1 becomes non-zero. Similar arguments hold for the field along any of the other three crystallographic directions in the basal plane. Evidence for this transition may already exist in the ac magnetic susceptibility measurements of Yoshida *et. al.* [11]. They observed a second peak in the imaginary part of the magnetic susceptibility only when the flux lines were parallel to the basal plane.

For a more detailed analysis consider the following dimensionless GL free energy density for the Γ_{5u} REP

$$\begin{aligned} f = & -|\vec{\eta}|^2 + |\vec{\eta}|^4/2 + \beta_2(\eta_1\eta_2^* - \eta_2\eta_1^*)^2/2 + \beta_3|\eta_1|^2|\eta_2|^2 + |D_x\eta_1|^2 + |D_y\eta_2|^2 \\ & + \kappa_2(|D_y\eta_1|^2 + |D_x\eta_2|^2) + \kappa_5(|D_z\eta_1|^2 + |D_z\eta_2|^2) \\ & + \kappa_3[(D_x\eta_1)(D_y\eta_2)^* + h.c.] + \kappa_4[(D_y\eta_1)(D_x\eta_2)^* + h.c.] + h^2. \end{aligned} \tag{1}$$

where $h = \nabla\times\mathbf{A}$, $D_\nu = \nabla_\nu/\kappa - iA_\nu$, f is in units $B_c^2/(4\pi)$, lengths are in units $\lambda = [\hbar^2c^2\beta_1/(16e^2\kappa_1\alpha\pi)]^{1/2}$, h is in units $\sqrt{2}B_c = \Phi_0/(4\pi\lambda\xi)^{1/2}$, $\alpha = \alpha_0(T-T_c)$, $\xi = (\kappa_1/\alpha)^{1/2}$, and $\kappa = \lambda/\xi$. Note that λ, ξ, B_c and κ are simply convenient choices and do not correspond to measured values of these parameters. A thorough analysis of Eq. 1 is difficult due to the unknown phenomenological parameters $\beta_2, \beta_3, \kappa_2, \kappa_3$, and κ_4. To simplify the analysis these parameters are determined in the weak-coupling, clean-limit for an arbitrary Fermi surface. Taking for the Γ_{5u} REP the gap function described by the pseudo-spin-pairing gap matrix: $\hat{\Delta} = i[\eta_1 v_x/\sqrt{\langle v_x^2\rangle} + \eta_2 v_y/\sqrt{\langle v_x^2\rangle}]\sigma_z\sigma_y$ where the brackets $\langle\rangle$ denote an average over the Fermi surface and σ_i are the Pauli matrices, it is found that $\beta_2 = \kappa_2 = \kappa_3 = \kappa_4 = \gamma$ and $\beta_3 = 3\gamma - 1$ where $\gamma = \langle v_x^2 v_y^2\rangle/\langle v_x^4\rangle$. Note that $0 \le \gamma \le 1$ and that $\gamma = 1/3$ for a cylindrical or spherical Fermi surface. These parameters agree with the cylindrical Fermi surface results of Ref. [12]. It is easy to verify that in zero-field $(\eta_1, \eta_2) \propto (1, i)$ is the stable ground state for all γ.

It is informative to determine the values of γ that are relevant to Sr_2RuO_4. LDA band structure calculations [13,14] reveal that the density of states near the Fermi surface are due mainly to the four Ru $4d$ electrons in the t_{2g} orbitals. There is a strong hybridization of these orbitals with the O $2p$ orbitals giving rise to antibonding π^* bands. The resulting bands have three quasi-2D Fermi surface sheets labeled α, β, and $\tilde{\gamma}$ (see Ref. [2]). The α and β sheets consist of $\{xz, yz\}$ Wannier functions and the $\tilde{\gamma}$ sheet of xy Wannier functions. In general γ is not given by a simple average over all the sheets of the Fermi surface. A knowledge of the pair scattering amplitude on each sheet and between the sheets is required to determine γ [9,15]. Recently, to account for the large residual density of states observed in the superconducting state, it has been proposed that either the xy or the $\{xz, yz\}$ Wannier functions exhibit superconducting order [9]. This model implies that that there are two possible values of γ; one for the $\tilde{\gamma}$ sheet (γ_{xy}) and one for an average over the $\{\alpha, \beta\}$ sheets ($\gamma_{xz,yz}$). A tight binding model indicates $\gamma_{xy} = 0.67$ and $\gamma_{xz,yz} = 0.11$ [16]. These values are sensitive to changes in the parameters of the tight binding model, however the qualitative result that $\gamma_{xy} > 1/3$ and $\gamma_{xz,yz} < 1/3$ is robust. Physically $\gamma_{xy} > 1/3$ because of the proximity of the $\tilde{\gamma}$ Fermi surface sheet to a Van Hove singularity and $\gamma_{xz,yz} < 1/3$ due to quasi 1D nature of the $\{\alpha, \beta\}$ surfaces [13,14].

Following Burlachov [17] for the solution of upper critical field $H_{c_2}^{ab}$ for the field in the basal plane, the vector potential is taken to be $\mathbf{A} = Hz(\sin\theta, -\cos\theta, 0)$ (θ is the angle the applied magnetic field makes with the $\hat{x}$ direction). After setting the component of $\mathbf{D}$ along the field to be zero it is found that $H_{c_2}^{ab}(\theta) = \kappa/(\kappa_5\lambda(\theta)/2)^{1/2}$ where $\lambda(\theta) = 1 + \gamma - [(1-\gamma)^2 - (1+\gamma)(1-3\gamma)\sin^2 2\theta]^{1/2}$. A measurement of the temperature independent four-fold anisotropy in $H_{c_2}^{ab}$ thus determines γ. To determine the field at which the second transition discussed above occurs consider the magnetic field along the $\hat{x}$ direction. The free energy of Eq. 1 is then similar to that studied in UPt_3 [18,19,10] and since Sr_2RuO_4 is a strong type II superconductor with a GL parameter of 31 for the field in the basal plane [11] the procedure of Garg and Chen [18] to study the second transition can be applied here. At $H_{c_2}^{ab}$ η_1 orders and and the vortex lattice solution is given by [10,18,19]

$$\eta_1 = \sum_n c_n e^{inqz} e^{-(\kappa_5/\gamma)^{1/2}\kappa H[y - qn/(\kappa H)]^2/2}. \tag{2}$$

where $c_n = e^{in^2\pi/2}$ and q has the two possible values $q_1^2 = \sqrt{3}H\kappa\pi(\gamma/\kappa_5)^{1/2}$ or $q_2^2 = H\kappa\pi(\gamma/\kappa_5)^{1/2}/\sqrt{3}$ (these two solutions are degenerate in energy). At the second transition the η_2 component becomes non-zero. As is discussed in Refs. [18,19] the solution for η_2 corresponds to a lattice that is displaced relative to that of η_1 by $\mathbf{d} = (\bar{y}, \bar{z})$. Accordingly, the field at which the second transition occurs is found by substituting

$$\eta_2 = ir\sum_n c_n e^{i(nq + \kappa H\bar{y})(z - \bar{z})} e^{-\sqrt{\kappa_5}\kappa H[y - \bar{y} - qn/(\kappa H)^2]/2} \tag{3}$$

and Eq. 2 into the free energy, minimizing with respect to the displacement vector $\mathbf{d}$, and determining when the coefficient of r^2 becomes zero. This yields for the ratio of the second transition (H_2) to the upper critical field

$$\frac{H_2}{H_{c_2}^{ab}} = \gamma^{1/2} \frac{\beta_A - \gamma(2S_1 - |S_2|)_{min}}{\beta_A - \gamma^{3/2}(2S_1 - |S_2|)_{min}} \tag{4}$$

where $\beta_A = 1.1596$, $S_1 = \overline{|\eta_1|^2|\eta_2|^2}/(\overline{|\eta_1|^2}\ \overline{|\eta_2|^2})$, $S_2 = \overline{(\eta_1\eta_2^*)^2}/(\overline{|\eta_1|^2}\ \overline{|\eta_2|^2})$, the over-bar denotes a spatial average, and the subscript min means take the minimum value with respect to $\mathbf{d}$ and with respect to $q = q_1$ or $q = q_2$. The numerical solution of Eq. 4 is shown in Fig. 1. Three vortex lattice configurations are found to be stable as a function of γ (depicted in Fig. 1). For $0 < \gamma < 0.187$ $q = q_2$ and $\mathbf{d} = (T_y, T_z)/4$ (T_y and T_z are the translation vectors of the centered rectangular cell for the η_1 lattice), for $0.187 < \gamma < 0.433$ $q = q_1$ and $\mathbf{d} = (T_y, T_z)/4$, and for $0.433 < \gamma < 1$ $q = q_2$ and $\mathbf{d} = 0$. For the field along $\hat{x} \pm \hat{y}$ the ratio H_2/H_{c_2} is given by Eq. 4 with γ replaced by $(1-\gamma)/(1+3\gamma)$. As a consequence the form of the vortex lattice will depend on the field direction. As has been discussed in detail in Ref. [20] the shape of the vortex lattice unit cell for $H < H_2$ will be strongly field dependent.

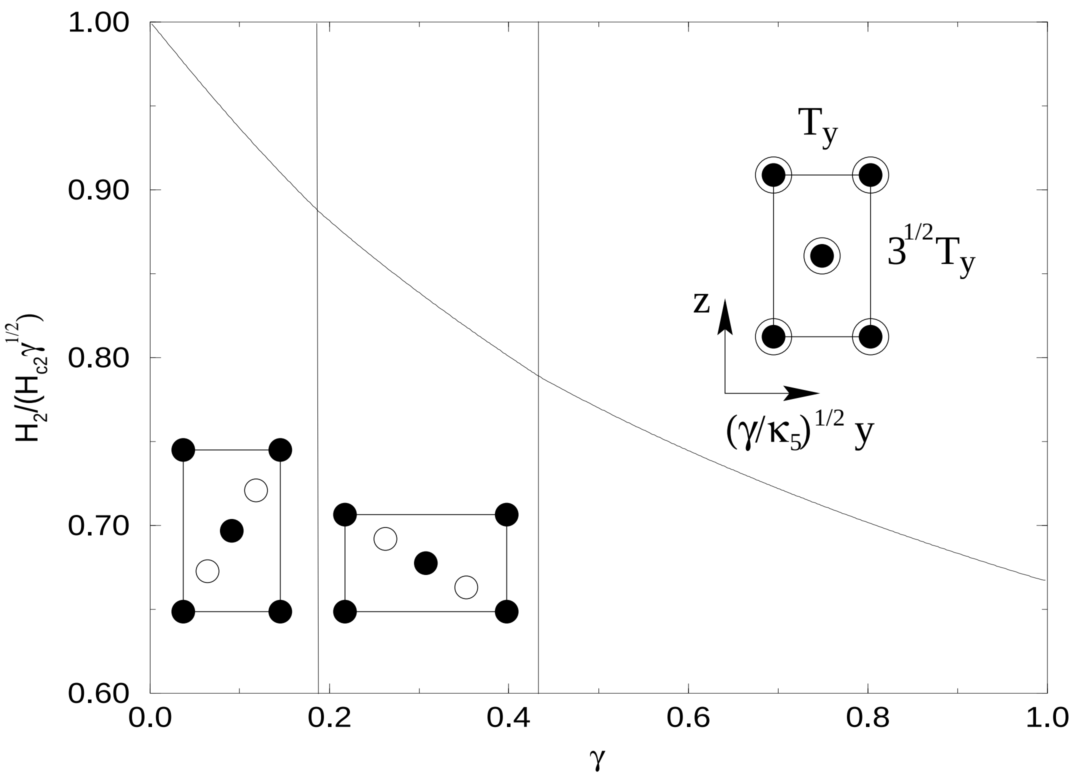

FIG. 1. The ratio of the two transition lines for the field along $\hat{x}$ as a function of γ. The open (closed) circles correspond to the zeroes of the η_2 (η_1) lattice. The vertical lines separate regions where the depicted vortex lattice structures are favored. For all three lattice structures the y and z axes have the same orientation and the dimensions of the rectangular cell are the same.

Consider the magnetic field oriented along the c-axis. Setting $D_z = 0$ writing $\Pi_+ = \kappa(iD_x + D_y)/(2H)$, $\Pi_- = \kappa(iD_x - D_y)/(2H)$, $\eta_+ = (\eta_x + i\eta_y)/\sqrt{2}$, and $\eta_- = (\eta_x - i\eta_y)/\sqrt{2}$, minimizing the quadratic portion of Eq. 1 with respect to η_+ and η_- yields

$$2\kappa \begin{pmatrix} \eta_+ \\ \eta_- \end{pmatrix} = H \begin{pmatrix} (1+\gamma)(1+2N) & (1+\gamma)\Pi_+^2 + (1-3\gamma)\Pi_-^2 \\ (1+\gamma)\Pi_-^2 + (1-3\gamma)\Pi_+^2 & (1+\gamma)(1+2N) \end{pmatrix} \begin{pmatrix} \eta_+ \\ \eta_- \end{pmatrix}. \tag{5}$$

where $N = \Pi_+\Pi_-$. The maximum value of H that allows a non-zero solution for (η_+, η_-) yields the upper critical field $H_{c_2}^c$. For $\gamma \neq 1/3$ $H_{c_2}^c$ must be found numerically (note that for $\gamma = 1/3$ the solution can be found analytically [21,8]). Expanding (η_+, η_-) in terms of the eigenstates of N (Landau levels) up to $N = 32$ and diagonalizing the resulting matrix yields the result for $H_{c_2}^c(\gamma)$ shown in Fig. 2. The solution for the form of the vortex lattice represents

a complex problem due to presence of many Landau levels in the solution of (η_+, η_-) and the weak type II nature of Sr_2RuO_4 for the field along the c axis (Ref. [11] indicates $\kappa \approx 1.2$). Here I present results that are strictly valid in the large κ limit and leave the treatment for general κ to a later publication (a perturbative calculation indicates that the qualitative results are unchanged for $\kappa = 1.2$) [16]. The form of the eigenstate of the $H_{c_2}^c$ solution is found to be $\eta_+(\mathbf{r}) = \sum_{n\geq 0} a_{4n+2}\phi_{4n+2}(\mathbf{r})$ and $\eta_-(\mathbf{r}) = \sum_{n\geq 0} a_{4n}\phi_{4n}(\mathbf{r})$ where $\phi_n(\mathbf{r}) = \sum_m c_m e^{iqm\tilde{y}} 2^{-n/2} H_n(\tilde{x} - qm/(\kappa H)) e^{-\kappa H(\tilde{x}-qm/(\kappa H))^2/2}/(n!)^{1/2}$ where the coefficients a_n are real, $(\tilde{x}, \tilde{y})$ is the vector (x, y) rotated by an angle $\tilde{\theta}$ about the z axis and $H_n(x)$ represent Hermite polynomials. In the large κ limit the form of the vortex lattice is found by minimizing $\beta = \overline{f_4}/(\overline{|\eta|^2})^2$ (f_4 is the quartic part of Eq. 1) with respect to the coefficients c_n, q, and $\tilde{\theta}$. It is assumed that $c_n = c_{n+2}$. This restricts the vortex lattice structures to be centered rectangular with a short axis $L_y = 2\pi/q$ and a long axis $L_x = 2q/(\kappa H)$. The ratio $t = L_x/L_y$ is $\sqrt{3}$ for a hexagonal vortex lattice and is 1 for a square vortex lattice. I further restrict the analysis to the two orientations $\tilde{\theta} = \{0, \pi/4\}$ since these correspond to aligning one of the vortex lattice axes with one of the high symmetry directions in the basal plane. Remarkably, the treatment of the many Landau levels in the solution of η_+ and η_- becomes numerically straightforward when β is expressed as a sum over the reciprocal lattice given by $\mathbf{l} = \hat{x}l_1 2\pi/L_x + \hat{y}l_2 2\pi/L_y$ [16] (see also Ref. [22]). It is found that β is minimized for $c_n = e^{in^2\pi/2}$ and that the values of t and $\tilde{\theta}$ depend upon γ. For $\gamma \leq 1/3$ ($\gamma \geq 1/3$) $\tilde{\theta} = 0$ ($\tilde{\theta} = \pi/4$) and t varies continuously from $\sqrt{3}$ to 1 as γ decreases (increases) from 1/3 to $1/3 - 0.0050$ ($1/3 + 0.0050$). For $\gamma < 1/3 - 0.0050$ and $\gamma > 1/3 + 0.0050$ the minimum β corresponds to $t = 1$. This implies that for $\gamma_{xz,yz}$ a square vortex lattice rotated $\pi/4$ about the c axis from the crystal lattice is expected and for γ_{xy} a square vortex lattice that is aligned with the underlying crystal lattice is expected near $H_{c_2}^c$. Note the appearance of the square vortex lattice correlates with an anisotropy in $H_{c_2}^{ab}$ of $|1 - H_{c_2}^{ab}(\theta = 0)/H_{c_2}^{ab}(\theta = \pi/4)| > 0.01$.

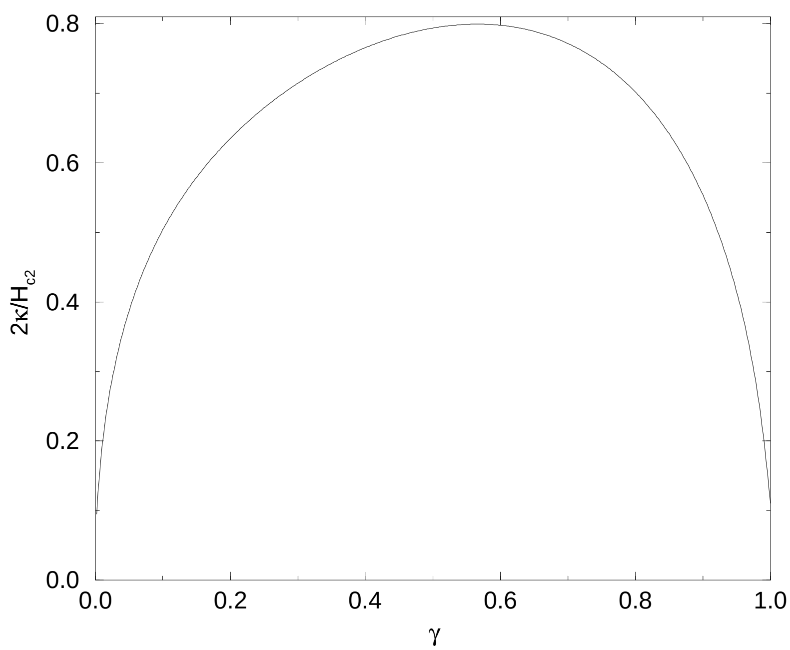

FIG. 2. The inverse of the upper critical field as a function of γ for the field applied along the c-axis.

The recent observation of a square vortex lattice in Sr_2RuO_4 [23] makes it of interest to compare the above behavior to that expected for the 1D REPS of D_{4h}. It is well known that for single component order parameters non-local corrections to the standard GL theory stabilize a square vortex lattice [24–26]. In particular the the following non-local term will stabilize the square lattice

$$\epsilon[|(D_x^2 - D_y^2)\psi|^2 - |(D_xD_y + D_yD_x)\psi|^2]. \tag{6}$$

Treating this term as a perturbation to the GL free energy leads to $\psi = \phi_0 - \tilde{\epsilon}\phi_4$ where $\tilde{\epsilon} = \sqrt{6}\epsilon H/\kappa$ (κ is the GL parameter) near $H_{c_2}^c$. As $\tilde{\epsilon}$ increases (note $\tilde{\epsilon} = 0$ at T_c) the vortex lattice continuously distorts from hexagonal to

square [24–26] until the square vortex lattice is stable for $|\tilde{\epsilon}| > 0.024$. The sign of ϵ determines the orientation of the vortex lattice; for $\epsilon > 0$ the vortex lattice is aligned with the underlying lattice while for $\epsilon < 0$ the lattice is rotated $\pi/4$ with respect to the underlying crystal lattice [24,26]. The sign of ϵ has been determined within a weak coupling clean limit approximation for the 1D odd-parity REPS. For the A_{1u} REP using $\hat{\Delta} = \psi(\hat{x}v_x/\sqrt{\langle v_x^2\rangle} + \hat{y}v_y/\sqrt{\langle v_x^2\rangle}) \cdot \vec{\sigma} i\sigma_y$ the sign of ϵ is determined by the sign of $3\langle(v_x^2+v_y^2)v_x^2v_y^2\rangle/\langle(v_x^2+v_y^2)v_x^4\rangle - 1$. Using a form for $\hat{\Delta}$ that is analogous to that used for the A_{1u} REP, the same result is found for all the 1D odd-parity REPS. This implies that the final orientation of the square vortex lattice for the $1D$ REPS is the same as that found for the $2D$ REP for superconducting order in the xy or the $\{xz, yz\}$ orbitals. The behavior of the vortex lattice for the 1D REPS as a function of $\tilde{\epsilon}$ is very similar to that for the 2D REP as a function of γ. An observable difference between the 2D and the 1D REPS is that for the 2D REP the vortex lattice remains square up to T_c while for the 1D REPS the vortex lattice is hexagonal at T_c. Also, the GL theories for the 1D and the 2D REPS predict a four-fold anisotropy in $H_{c_2}^{ab}$ but this anisotropy vanishes at T_c for the 1D REPS and does not vanish at T_c for the 2D REP.

In conclusion I have examined GL models for the odd-parity REPS of the superconducting state for Sr_2RuO_4. It was found that if the zero-field ground state breaks $\mathcal{T}$ symmetry (the 2D REP) then there should exist a second transition in the mixed state when the magnetic field is applied along a high symmetry direction in the basal plane. It was also shown that when the field is along the c-axis there will be a square vortex lattice for all the possible odd parity superconducting states.

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